COPRECIPITATION OF RADIUM

WITH METAL ARSENATES

by

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SUMMARY

This report describes the coprecipitation of lead, thorium and radium-226 as arsenates over the pH range of one to twelve. The coprecipitating cations are zinc, copper, ferric iron and barium. Lead is precipitated with all cations from a pH of 4 to a pH of 12. Thorium can be precipitated as an arsenate over the total pH range as well as from a ten molar acid solution.

Radium arsenate is similar to barium arsenate, it is highly insoluble, showing slight solubility around a pH of 2.

A second phase of this project was concerned with chemical barriers to radium, thorium and lead in aqueous solutions. A nine inch barrier of Elliot Lake tailings, 1% in barium arsenate or ferric arsenate, was sufficient to prevent the passage of radium, thorium and lead in a synthetic solution. Thorium and radium were reduced to a non-detectable level at pH's of 2 and 6. Lead was removed at pH of 6, whereas at a pH of 2 only 20% was removed.

COPRECIPITATION OF RADIUM WITH METAL ARSENATES

INTRODUCTION

The first plant to treat uranium ore was built in Czechoslovakia in 1906. It wasn't until 1913 that a second plant was built in Denver, Colorado. In Canada, Eldorado Mining and Refining Ltd began working the uranium deposits at Great Bear Lake in 1933. These early operations were primarily concerned with the radium content of the ores.

The uranium industry did not exist in 1940, previous to this time uranium was a by-product of the radium industry. Following World War II the development of nuclear power provided a demand for uranium, hence the expansion of mining and processing of uranium ores.

The mining and milling of uranium ores, by 1982, had produced 133,000,000 tons of uranium tailings at the rate of 25,000 - 30,000 tons daily. Uranium ores contain a number of radionuclides which are inherently dangerous and are associated with the tailings after separation of the uranium from the ore. The primary radioactive contaminants associated with tailings are thorium, radium, radon, lead and polonium.

Wastes, which make up tailings, include wash water, sands, waste solids and various process liquors. These wastes are stored in tailings ponds or lagoons where the volume is reduced by evaporation and seepage of the liquid portion. The composition of tailings in the Elliot Lake area is given in Table 1. These analyses are for the various factions which make up the tailings pond.

TABLE 1

COMPOSITION OF ELLIOT LAKE TAILINGS

	Slimes	Sands
I. Tailings Solids		
Minerals (relative Concentrations		
- Gypsum (%)	10-12	1-4
- Carbonate (%) (Calcite)	0.2	0.005
- Iron (%) ($Fe(OH)_3$)	3	0.5
- Pyrite (FeS_2)	3-7	3-7
Radionuclides		
- Ra-226 (pCi/g)	100-500	100-200
- Pb-210 (pCi/g)	100	100
- Po-210 (pCi/g)	50	50
- U (ug/g)	10-100	10-100
- Th (ug/g)	10-300	10-300
Other Constituents		
- Fe	-	-
- Cu	-	-
- Zn	-	-
- Ni	-	-
- Co	-	-
- Pb	-	-
- As	-	-
II. Pore Water Chemistry		
Radionuclides		
- Ra-226(pCi/L)	10-300	10-300
- Pb-210 (pCi/L)	5-50	50-5,000
- Po-210 (pCi/L)	-	-
- U (ug/L)	50-200	100-5,000
- Th (ug/L)		
pH		
- discharge	9-11	9-11
- weathered	3-6	1-5

TABLE 1 (Continued)

COMPOSITION OF ELLIOT LAKE TAILINGS

	Slimes	Sands
Other Constituents (mg/L)		
- Al	1-50	1-200
- SiO_2	20-120	20-120
- Ca	400-600	400-600
- Mg	10-250	10-400
- Na	10-40	10-40
- K	10-130	10-170
- SO_4	1,500-10,000	10,000-150,000
- PO_4	0-2	0-2
- Fe	50-5,000	1,000-30,000
- Cu	0-5	0-10
- Zn	5-20	1-50
- Ni	1-5	1-30
- Co	0-4	1-30
- Pb	0-0.5	0-4
- Mn	20-200	0-30

REFERENCE: Beak Consultants Limited, Waste Management Implications of Concentrating Slimes - Characteristics and Potential Problems Report for Atomic Energy Control Board, Ottawa, Canada, May 1986.

During the 1950's and 1960's tailings were deposited as slurries, 30 to 40% solids, in settling ponds for sedimentation of solids without water treatment for the precipitation of radium-226. These settling ponds were constructed by damming low areas in the vicinity of the uranium mill. Radium removal from tailing basin effluents has been common practice since 1965. The treatment consists of adding barium chloride to precipitate barium - radium sulphate which is not completely efficient as it allows suspended radium-226 to be released.

In 1974 a task force was established to develop effluent regulations and guidelines for the uranium mining industry. One of the recommendations of this task force was the need to seek more effective methods of recovering radium-226 from tailings so as to reduce the radium-226 content of uranium mining and mill effluents to 10 pCi/L total and 3 pCi/L dissolved.

Other means of impounding uranium mine and mill tailings are in use or have been suggested. In Saskatchewan, Gulf uses a pit disposal method in which the pit is an old mine. Key Lake uses a sub-aerial deposition scheme in which the fine tailings cover the coarse tailings. This blanket of fine tailings prevents the water from reaching the coarse tailings underneath as it acts like a clay. This clay material is also effective as a barrier to radon gas.

The disposal of tailings in deep lakes has also been suggested. The lake water would act as a blanket preventing exposure of the material to air preventing oxidation and acidification of the tailings.

Effluents from base metal, uranium and iron ore mines are subject to controls to limit the discharge of deleterious substances. These controls are expected to provide protection to fish and other aquatic life. Limits have been specified for those parameters which are known to occur commonly in mining effluents in sufficient amounts to be deleterious to fish. The parameters and their limits are summarized in Table 2.

TABLE 2

SUMMARY OF ACCEPTABLE EFFLUENT LEVELS AS SET OUT IN REGULATIONS AND GUIDELINES

(For Base Metal, Uranium and Iron Mines)

PARAMETER	CONTROL LEVEL* (TOTAL METALS)
Lead	0.1 mg/1
Copper	0.3 mg/1
Arsenic	0.5 mg/1
Nickel	0.5 mg/1
Zinc	0.5 mg/1
Total Suspended Matter	25 mg/1
pH	6.0 minimum
Acute Lethality	Not less than 50% survival in a 96-hr acute lethality test (in guidelines only)
Ra 226**	10 pCi/1 (dissolved)

* The control level represents a monthly arithmetic mean for the parameter listed.

** For Ra 226 the control level refers to a dissolved analysis using a 3 micron filter.

The majority of the uranium mills in Canada use a sulphuric acid leach process to extract the uranium producing acidic residues and acidic liquid wastes. These wastes must be treated prior to discharge of the effluent water to minimize their impact on the environment and meet the guideline limits given in Table 2 for effluent quality. The treatment of acid wastes consists of neutralization with lime to a pH of 8 to 11.5 precipitating the heavy metals. The solid and liquid phases are separated in a settling pond. The liquid phase is then treated with $BaCl_2$ to precipitate the radium-226 as a barium radium sulphate compound in settling tanks. The resulting water is discharged

to the environment. The concentration of radium-226 in the discharge water is usually in the vicinity of 2 to 3 pCi.

This study is concerned with the chemistry of radium arsenate to ascertain if by the treatment of uranium mill effluents with arsenate a more stable tailings area will result with a lower concentration of radium-226 in the discharge waters. Tailings, which contain pyrite, become acidic with time through the production of sulphuric acid by sulphide oxidation hence the variation of radium arsenate precipitation as a function of pH with various coprecipitants is a parameter to know before applying arsenate for the treatment of uranium mill tailings. Furthermore, it is essential to determine the effect of arsenate and pH on the other radionuclides associated with uranium ores, i.e. lead and thorium.

THEORY

The immobilization of radioactive elements such as radium, thorium and lead requires that these elements form stable and highly insoluble compounds. Sulphate is presently used with barium as a coprecipitant in immobilizing radium. A comparison of solubilities of the sulphates of barium and radium with the solubilities of their arsenates indicates that arsenate has a thousand fold advantage over sulphate as a reagent to immobilize radium.

The solubility product of divalent metals with arsenate is represented in equation 1.

$$K_{SP} = [M]^3 [AsO_4]^2 \quad (1)$$

where M represents the divalent metal ion.

Solubility products are expressed in terms of the reacting ions only, hence without considering hydrolysis of the ions they have unique solutions. Considering hydrolysis the concentrations of the reactants varies with pH due to the formation of hydroxylated metal ions and protonated arsenate species. The dissociations and formations of the various ions are depicted by equations 2 to 8.

$$H_3AsO_4 \rightleftharpoons H_2AsO_4^- + H^+ \quad (2)$$

$$H_2AsO_4 \rightleftharpoons HAsO_4 + H^+ \quad (3)$$

$$HAsO_4 \rightleftharpoons AsO_4 + H^+ \quad (4)$$

$$M^{++} + OH^- \rightleftharpoons MOH^+ \quad (5)$$

$$M(OH)^+ + OH^- \rightleftharpoons M(OH)_2 \quad (6)$$

$$M(OH)_2 + OH^- \rightleftharpoons M(OH)_3^- \quad (7)$$

$$M(OH)_3^- + OH^- \rightleftharpoons M(OH)_4^= \quad (8)$$

The total concentrations can thus be expressed as the sums of the individual terms as in equations 9 and 10.

$$[M_t] = [M^{++}] + [M(OH)^+] + [M(OH)_2] + [M(OH)_3^-] + [M(OH)_4^=] \quad (9)$$

$$[As_t] = [H_3AsO_4] + [H_2AsO_4^-] + [HAsO_4^=] + [AsO_4^{\equiv}] \quad (10)$$

Relating the individual terms to their formation and dissociation constants the total ion concentrations can be expressed as functions of the reacting species, total metal and arsenate concentration, hydrogen ion concentration and their formation and dissociation constants. These derived expressions are given by equations 11 and 12.

$$[M_t] = [M^{++}]\,[1 + [OH]K_1 + [OH]^2K_1K_2 + [OH]^3K_1K_2K_3 + [OH]^4K_1K_2K_3K_4] \quad (11)$$

$$[As_t] = [AsO_4]\left[1 + \frac{[H^+]}{K_3} + \frac{[H^+]^2}{K_2K_3} + \frac{[H^+]^3}{K_1K_2K_3}\right] \quad (12)$$

The terms in brackets containing the dissociation constants have been disignated as β_m and β_{As} for ease of handling. Substituting these values into equation 1, equation 13 is obtained expressing the solubility of $M_3(AsO_4)_2$ in terms of total concentrations of ions in solution and hydrogen ion concentrations.

$$K_{SP} = \frac{[M_t]^3}{(\beta_m)^3} \cdot \frac{[As_t]^2}{(\beta A_s)^2} \quad (13)$$

The dissolution of a salt $M_3(AsO_4)_2$ in water is given by equation 14.

$$M_3(AsO_4) \rightleftharpoons 3M^{++} + 2\ AsO_4^{\equiv} \quad (14)$$

Thus, for S moles of $M_3(AsO_4)_2$ dissolved in 1 litre of solution there will be 3S moles of M and 2S moles of $AsO_4^{\equiv}$, hence substituting these values into equation 13, equation 15 is derived.

$$K_{sp} = 108\ S^5/\beta_m 3 \cdot \beta_{As}{}^2 \qquad (15)$$

Solving this equation for S at various pH's provides theoretical solubility values for various metal arsenates.

Equation 15 has been solved for the arsenates of lead, copper, zinc and barium using the solubility products in Table 3. The results of these calculations are given in Figure 1.

Equation 15 reduces to equation 15a for ferric arsenate. The solubility of ferric arsenate as the concentration of ferric iron is also given in Figure 1.

$$K_{SP} = S^2/\beta_m \cdot \beta A_s \qquad (15a)$$

The theoretical solubilities of the metal hydroxides are given in Figure 2. These results were calculated from the cumulative formation constants of the metal hydroxy complexes, Table 4, and the solubility products of the hydroxides in Table 4. The general equation for these calculations is:

$$[M] = K_{SP} \cdot \beta/[OH]^2 \qquad (16)$$

Assuming the solubility product of radium arsenate is similar to the solubility product of barium arsenate (8.0×10^{-51}) then the solubility of radium arsenate as a function of pH would be practically the same as that for barium shown in Figure 1. Figure 1 shows that barium arsenate, hence radium arsenate, is much less soluble over the entire pH range than any of the other arsenates. It is thus expected that radium can be coprecipitated with zinc, copper, ferric iron and barium as the arsenates.

TABLE 3

SOLUBILITY PRODUCTS OF BARIUM, ZINC, LEAD, COPPER AND FERRIC IRON HYDROXIDES AND ARSENATES *

METAL	SOLUBILITY PRODUCTS	
	HYDROXIDES	ARSENATES
Ba	5×10^{-3}	8.0×10^{-51}
Zn	1.2×10^{-17}	1.3×10^{-28}
Pb	1.1×10^{-20}	4.0×10^{-36}
Cu	2.2×10^{-20}	7.6×10^{-36}
Fe (111)	4×10^{-38}	5.7×10^{-21}
Th	4×10^{-45}	

* Dean, John A., Langes Handbook of Chemistry, 13th Ed., McGraw-Hill Book Company, New York, 5-6.

TABLE 4

CUMULATIVE FORMATION CONSTANTS OF METALS WITH HYDROXIDE*

METAL	K_1	K_2	K_3	K_4
Barium				
Copper	1×10^{7}	4.8×10^{13}	1×10^{17}	3.3×10^{18}
Iron (111)	7.5×10^{11}	1.5×10^{21}	4.7×10^{29}	
Zinc	2.5×10^{4}	2.0×10^{11}	1.4×10^{14}	4.6×10^{17}
Lead	6.6×10^{7}	7.1×10^{10}	3.8×10^{14}	
Th	7.8×10^{3}	1.3×10^{4}		

* Dean, John A., Langes Handbook of Chemistry, 13th Ed., McGraw-Hill Book Company, New York, 5-14.

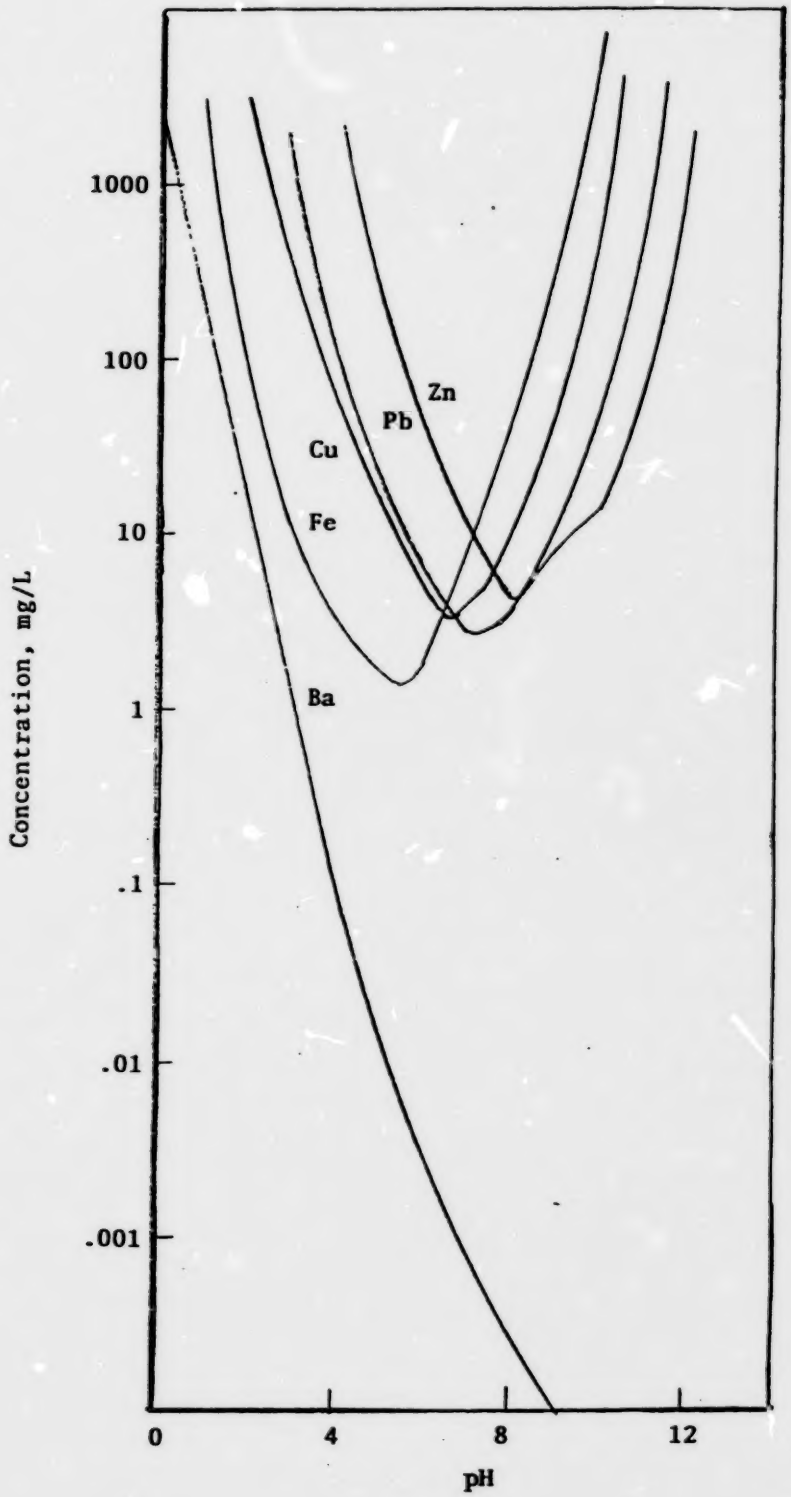

Figure 1 Theoretical Solubilities of Metal Arsenates as a Function of pH

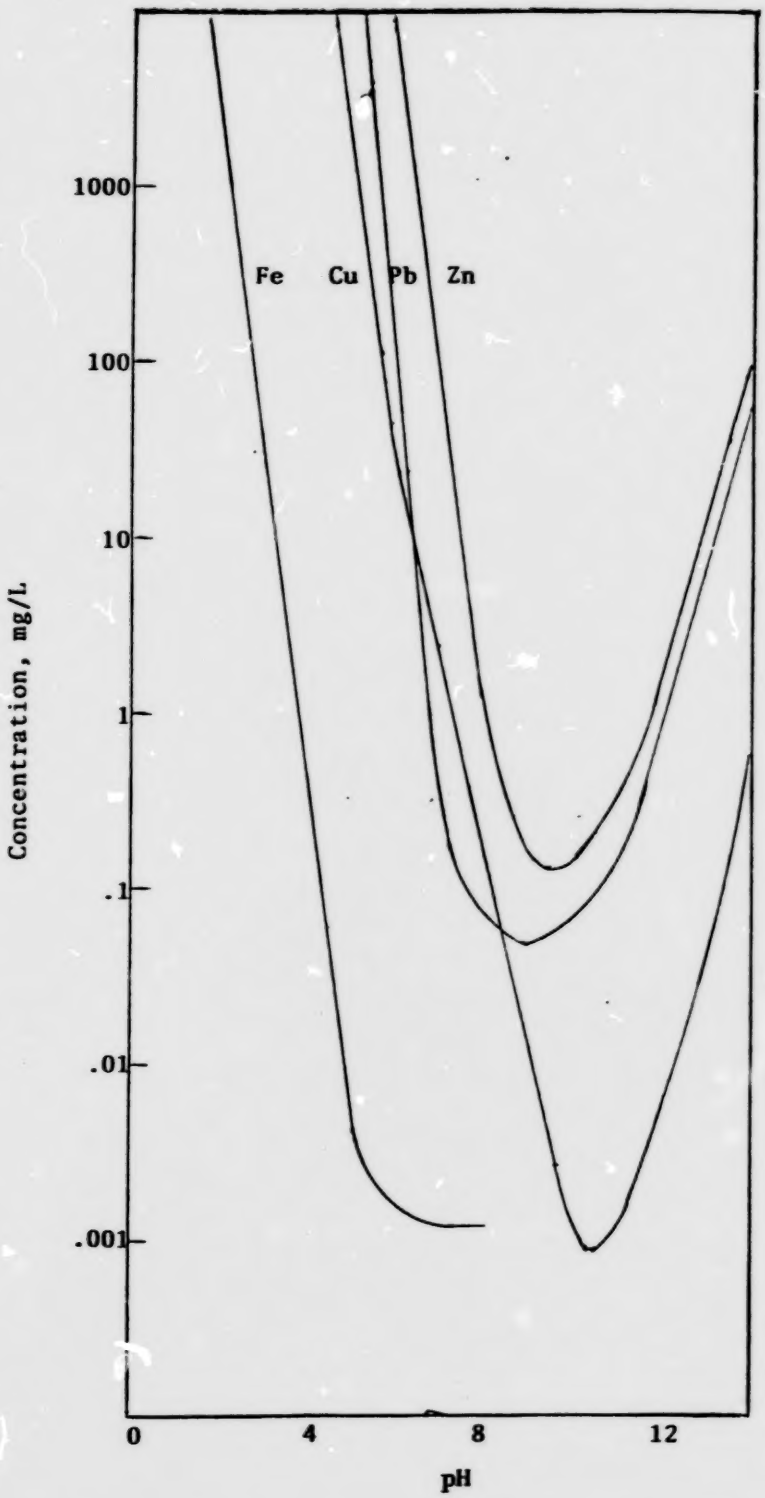

Figure 2 Theoretical Solubilities of Metal Hydroxides as a Function of pH

COPRECIPITATION

I. Experimental Methods

The Elliott Lake tailings were supplied by Energy, Mines and Resources Canada. All other chemicals were from commercial sources. The preparation of solutions and analytical methods are described in the Appendix.

A systematic investigation of the variables which affect solubility of the arsenates of the radioactive elements lead, thorium and radium was accomplished by coprecipitation from solution of different pHs over the pH range of 1 to 12. The coprecipitants were zinc, copper, barium and ferric iron at three different concentrations. The variations in solubility with change in pH were followed by analyzing the supernatant liquors for the radioactive component and the coprecipitated cation. These solubility studies were carried out in sulphate free and 0.75 M sulphate solution. The high sulphate solution studies were necessary as uranium mine with tailings contain sulphate added in the process.

The use of arsenate as a barrier to the transport of radium from a tailings area to the surrounding environment was investigated. A bed of the arsenate was prepared and transferred to a column through which was percolated an aqueous solution of the radioactive component. The concentration of component in the effluent was determined. The variables investigated in this system were pH, concentration and thickness of barrier.

II. Results and Discussions

A. Coprecipitation of Lead

The results of the coprecipitation experiments are tabulated in the Appendix. The solubility of lead as a function of pH is also presented graphically with the tables.

The coprecipitation of lead with zinc arsenate is reproduced in Figure 3 which contains the variation of lead concentration

as a function of pH for three different concentrations of zinc and arsenate. Curve A is a plot of the lead concentration in which the stoichiometric amount of arsenate is less than that required to react with the lead. This curve shows a decrease in lead concentration beginning at a pH of 3 and levelling off at a pH of 6. This level portion of the curve signifies the completion of the reaction between lead and arsenate. As the pH is increased beyond six the decrease in lead concentration is due to the formation of lead hydroxide.

Curves B and C are derived from data in which there is an excess of arsenate. With Curve B there was a onefold excess, in terms of lead, of arsenate over that required to precipitate the lead and zinc. Curve C was obtained with a threefold excess of arsenate in terms of lead. Curves B and C are very similar showing a minimum concentration of lead in the pH range of 4 to 8. The increase in concentration of lead above a pH of 8, is due to the formation of lead hydroxo complexes.

Results similar to the results with zinc arsenate were obtained with copper and barium as shown by the data in Figures B2 and B3 given in the Appendix. These curves show the same pH (3) for initial precipitation and the same pH range for minimum solubility. The similarity of these results is influenced by the same stoichiometry of the reactions, i.e. in all cases the metal to arsenate molar ratio is 3 to 2.

Ferric iron reacts with arsenate in the ratio of 1 to 1. The results of precipitating lead with ferric arsenate are plotted in Figure B4 in the Appendix. The resulting curves are similar to those with zinc, copper and barium except the range of minimum lead solubility is smaller, from 4 to 6.

Coprecipitation of lead was also carried out in a 0.75 M sodium sulphate solution. The results for zinc arsenate, copper arsenate, barium arsenate and ferric arsenate are given in Table B13 of the Appendix. The formation of lead sulphate is evident by the low concentrations of lead in the low pH range of

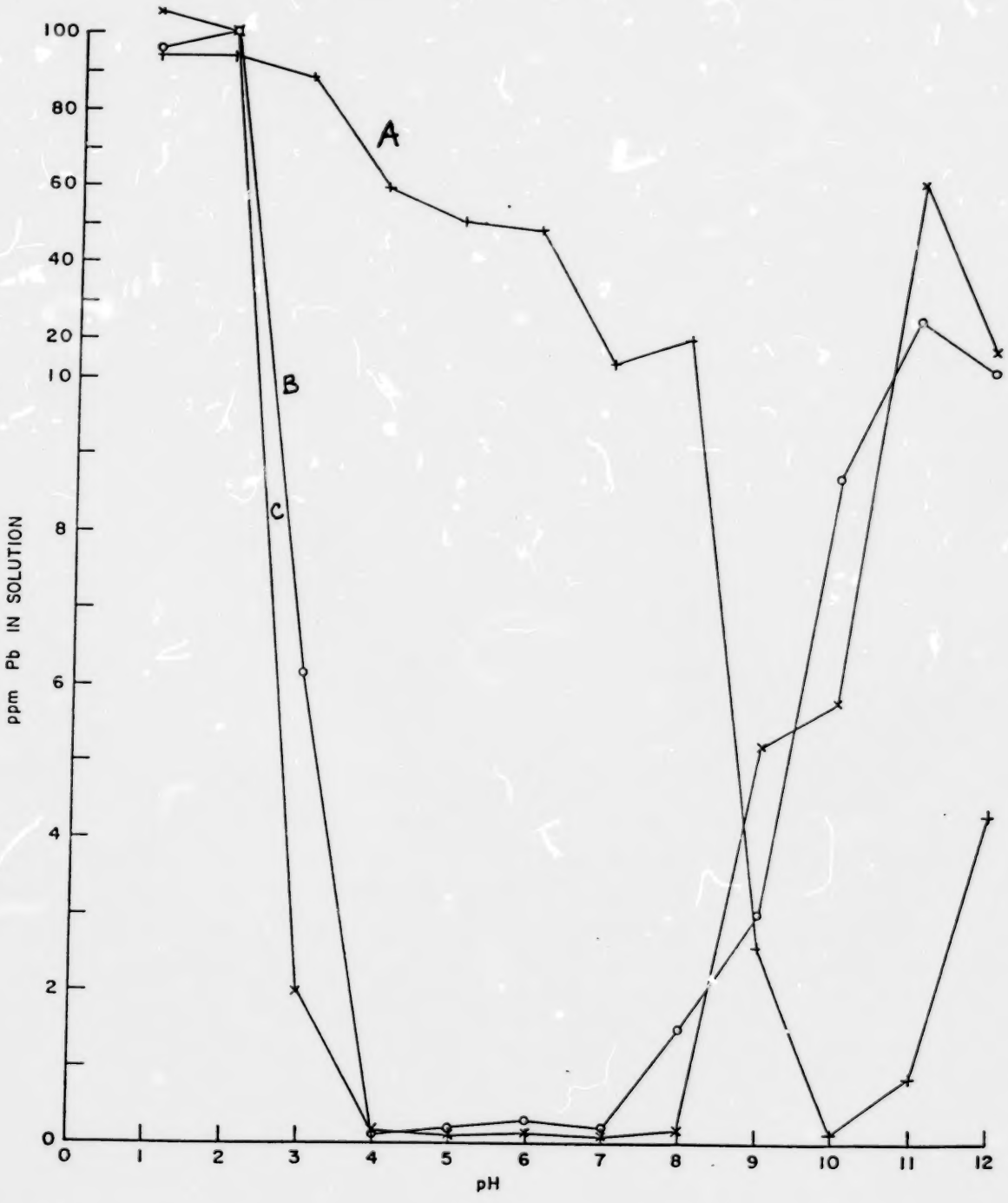

Fig. 3 Coprecipitation of Lead with Zinc Arsenate as a function of pH

Initial concentrations :

Pb	100 ppm
Zn	0.01 M + ; 0.1 M o ; 0.2 M ×
Arsenate	0.02 M + ; 0.2 M o ; 0.4 M ×

1 to 4. The effect of arsenate does not become evident until the acidity is decreased to a pH of 5 with the exception of barium arsenate which shows no influence of arsenate.

B. Coprecipitation of Thorium

The data for the coprecipitation of thorium is presented in the Appendix in Tables B5, B6, B7 and B8. Thorium behaved similarly with all four coprecipitants that is, with concentrations of 1×10^{-3} M and 2×10^{-3} M arsenate, thorium was completely removed from solution at a pH of 2 and remained insoluble up to a pH of 12. Coprecipitation experiments, in which the amount of arsenate was less than the stoichiometric quantity required to react with the thorium, yielded precipitation of thorium, concentrations of thorium less than 0.01 ppm, beginning at a pH of 6. These results correspond to the precipitation of thorium hydroxide and indicating that the low concentrations of thorium at lower pH's and higher concentrations of arsenate is due to the formation of thorium arsenate. Precipitation of thorium arsenate at acidities greater than pH = 2 is possible by increasing the arsenate concentration. This is shown in Table 5 which contains results in which the arsenate concentration is 2.5 times that required to react with the thorium and the acidity has been increased to 1 and 10 molar hydrogen ion solution.

TABLE 5

PRECIPITATION OF THORIUM FROM STRONG ACID SOLUTIONS

Acidity	Metal Present	Thorium in Solution
1 M	Fe	<0.01 ppm
	Cu	<0.01 ppm
10 M	Fe	<0.01 ppm
	Cu	<0.01 ppm

Initial Concentrations:

Thorium	100 ppm
Copper	1×10^{-3} M
Iron	1×10^{-3} M
Arsenate	4×10^{-3} M

The precipitation of thorium as a thorium arsenate from strong acid solution suggested that arsenate can be used to contain thorium in uranium mill tailings dumps. This containment is possible over the range of acidities generated by sulphide oxidation. This reaction also suggests that thorium can be recovered from acid uranium mill streams. Thorium arsenate is precipitated and filtered from solution as thorium arsenate. Other thorium compounds may then be produced by reduction of the arsenate to arsenite with dissolution of the thorium, separation and crystallization of the desired product.

Table B14 contains the results for the coprecipitation of thorium as the arsenate in a sulphate solution. Comparison of these results with the results from a non-sulphate solution indicates that the presence of sulphate enhances the precipitation of thorium arsenate. The pH for removal of thorium from solution has been extended from 2 to 1.

C. Coprecipitation of Radium

The data for the coprecipitation of radium is tabulated in the Appendix in Tables B9, B10, B11 and B12. The data for the coprecipitation of radium in the presence of sulphate is given in Table B15.

The data for the coprecipitation of radium as the arsenate shows that radium arsenate is precipitated over the pH range of 1 to 12. The percent radium removed from solution increases as the pH increases which corresponds to an increase in the formation of the metal arsenate, the coprecipitant. The increase in quantity of radium removed with an increase in quantity of coprecipitant formed is also shown by the increase in radium removed by increasing the quantity of reagents.

Comparing the results of coprecipitating radium with barium arsenate and ferric arsenate from a 0.75 M sulphate solution with the corresponding results from a non-sulphate solution shows that sulphate enhances the removal of radium from solution. Barium and ferric arsenate removed about 99% of the radium from a solution containing 500 pCi of radium. This reduction occurred over the pH range of 2 to 12. Arsenate, as an important factor in the removal of radium, is demonstrated by the similarity of the barium and iron results. In the case of barium a precipitate of barium sulphate is possible carrying radium sulphate, due to the solubility of ferric sulphate the corresponding coprecipitation with ferric iron is impossible, hence, ferric arsenate carries down the radium.

Moffett, D. and E. Barnes* showed that the efficiency of radium-226 removal by means of sulphate falls dramatically above a pH 9.5. Arsenate on the other hand is shown to sustain less than 1 pCi/L of soluble radium-226 at greater than pH 9.5 which is an advantage over sulphate.

* Moffett, D. and E. Barnes, Canadian Nuclear Association, 19th Annual International Conference, June 10 - 13, 1979 Proceedings.

RADIUM BARRIER

The results of the solubility experiments have shown that lead, thorium and radium are coprecipitated with the arsenates of zinc, copper, barium and ferric iron. These results are due to the insolubility of the radioactive nucleotides compared to the coprecipitants thus suggesting that the arsenate may be exchanged from the coprecipitant to the radioactive cation, thus removing the lead, thorium and radium from solution. The use of the arsenates of barium and iron as barriers to lead, thorium and radium were tested. These arsenates should act as ion exchange media and their effectiveness a function of pH, thickness of the barrier and the metal arsenate.

I. Method

The barriers were constructed in the form of columns through which solutions of radium were gravity fed. The effluents were analyzed for concentration of radium. The various barriers tested were uranium mill tailings and barite mill tailings treated with arsenate, barium arsenate and ferric arsenate mixed with uranium mill tailings. The ratio of mill tailings to the arsenates were varied and the pH at which these barriers were effective in removing radium was also studied.

II. Results and Discussion

The solubility data for the arsenates of the coprecipitants zinc and copper show that these arsenates begin to dissolve in the pH range of 5 to 6, hence, in tailings they are not applicable as barriers to radium in areas having low pHs. Barium and ferric iron on the other hand are more stable in more acidic conditions, hence, they were investigated as reactive species in forming a barrier to radium transport from uranium mill tailings dumps.

Table 6 contains the results from various barrier systems and a control column of tailings only for comparison. In all cases the pH of the effluent from these columns was 6 even if the pH of the influent was 3. This adjustment of pH was due to the presence of lime and limestone used in adjusting the pH of the tailings stream.

Uranium mine and mill tailings and barite mill tailings which have reacted with arsenate did not function very well as radium barriers. The untreated uranium mill tailing was not effective in removing radium and the barite tailings (0.15% As) mixed with uranium mill tailings did not completely remove radium until they were mixed in the ratio of 2 parts barite to 10 parts uranium mill tailings. The study of the use of these systems was terminated due to the high concentration of barite required for effective removal of radium.

Ferric hydroxide has been used to remove arsenic from domestic water supplies. The product in this process is ferric arsenate supported on an absorbent clay. This material was tested for removing radium from solution. The four inch barrier of this material, 0.12% in arsenic, reduced radium from 50 pCi/L to a non-detectable level, i.e. less than 1 pCi/L.

Uranium mill tailings containing barium arsenate or ferric arsenate have been found effective in removing radium from solution over the pH range of 2 to 6.6. The results of these experiments are given in Tables 6 and 7 in which the barium and ferric arsenates are mixed with uranium mill tailings in the ratios of 1 to 10 and 1 to 100. At the lower pH values the removal of radium is incomplete, only 95% being removed from a solution containing 1000 pCi per liter. These results were obtained with a 4 inch barrier. A 9 inch barrier of 1 part barium or ferric arsenate mixed with 100 parts uranium mill tailings reduced the radium concentration to an undetectable level at the low pHs.

TABLE 6

BARRIER LAYERS FOR RADIUM CONTAINMENT

Barrier	pH	pCi/L Ra-226 In Effluent	pCi/L Ra-226 In Effluent	Flow Rate ml/min
Control Tailings	6	10	10	10
	6	10	10	15
	6	50	50	10
Mill Tailings	6	10	10	15
Arsenate	6	50	20	10
Barite Tailings				
0.15% As With 1:00	6	50	20	10
Mill Tailings 2:10	3.0/5.6	50	0	10
1:10	6	50	10	10
Mill Tailings - Barium Arsenate				
	3.0/4.7	50	0	10
10:1	6	50	0	10
100:1	6	50	0	10
100:1	3.0/5.9	50	0	10
Spent Arsenic	6	50	0	700
Filter Packing	6	50	0	10
0.12% As	3.0/7.6	50	0	10
Mill Tailings - Ferric Arsenate				
10:1	6	50	0	10
100:1	3.0/6.6	50	25	10

TABLE 7

BARIUM AND FERRIC ARSENATES AS BARRIERS TO RADIUM

Column 4 inches
$Ba_3(AsO_4)_2$ + Mill Tailings 1:100

pH In/pH Out	pCi Ra^{226}/L Influent	pCi Ra^{226}/L Effluent	Flow Rate
6/	50	0	10
3.0/5.9	50	0	10
3.0/3.5	50	0	10
3.0/3.3	1000	0	10
2.0/2.0	1000	25	10
3.0/3.4	1000	9	10
3.0/3.7*	1000	0	10

* 9 Inch Column

$FeAsO_4$ + Mill Tailings 1:100

pH In/pH Out	pCi Ra^{226}/L Influent	pCi Ra^{226} Effluent	Flow Rate
3.0/6.6	50	25	10
3.0/3.6	50	0	10
3.0/3.6	1000	0	10
2.0/2.0	1000	49	10
3.0/3.7	1000	18	10
3.0/3.7*	1000	0	10

* 9 Inch Column

A 9 inch barrier system was tested to remove radium, thorium and lead. The results of these experiments are given in Table 8. At pH's of 6 and 2 radium and thorium concentrations were reduced to a non-detectable level. Lead, on the other hand, is reduced to a low level at the high pH but shows dissolution at pH of 2. The dissolution of lead at pH less than 4 is expected from the results of

the solubility studies. Lead, thorium, barium and iron also behave as demonstrated by the solubility results.

The stability of these layers to changes in pH is demonstrated by the low concentration of barium and ferric iron eluted from the columns. It is demonstrated that the barium forms a more stable barrier than ferric iron due to barium arsenate being less soluble than ferric arsenate.

TABLE 8

ARSENATE LAYERS AS BARRIERS TO RADIUM, THORIUM AND LEAD

9" Barrier
Flow Rate 10 ml/min
Influent: 1000 pCi/L Ra^{226}
10 mg/L Th
10 mg/L Pb

Barrier	pH In/Out	Effluent Ra	Effluent Th	Effluent Pb	Barrier Dissolution
9" $Ba_3(AsO_4)_2$	2/2	0	<0.01 ppm	4.5 ppm	<0.1 ppm Ba
	6/6	0	<0.01 ppm	0.1 ppm	<0.1 ppm Ba
9" $FeAsO_4$	2/2	0	<0.01 ppm	8.0 ppm	8.2 ppm Fe
	6/6	0	<0.01 ppm	0.3 ppm	0.4 ppm Fe

CONCLUSIONS

1. Arsenate precipitates radium and thorium from acid solution.

2. Arsenate precipitates lead from solutions at greater than pH 4.

3. Barium arsenate mixed with uranium mill tailings in the ratio of 1:100 forms an excellent barrier to the passage of radium, thorium and lead in acidic solutions.

4. Ferric arsenate mixed with uranium mill tailings is also a good barrier to radium, thorium and lead but does not retain lead at low pH's and it is not as stable as barium arsenate.

5. The presence of barium would prevent arsenic from becoming an environmental contaminant when used for this purpose.

6. A four inch barrier has treated water equivalent to a 6 year rainfall in the Elliot Lake area, hence a thicker barrier with a higher concentration of arsenate should contain radium, thorium and lead for a number of decades.

RECOMMENDATIONS

1. Barium arsenate replace barium sulphate to coprecipitate radium in uranium mill tailings streams.

2. Barium arsenate be used as a grout curtain on the periphery of old uranium tailings dump to contain radium, thorium and lead.

3. Before use of ferric or barium arsenate as a barrier the following factors as they effect the efficiency of the barrier system should be addressed:

   a) Wet-dry cycling
   b) Freeze-thaw cycling
   c) Aging
   d) Method of preparation
   e) Bacteria

APPENDIX A

1. Preparation of Solutions

Lead Nitrate, 400 ppm pb

Lead nitrate was prepared by dissolving 1.2790 g of $Pb(NO_3)_2$ in distilled water in a 2 L volumetric flask. After dissolution the solution was diluted to the mark.

Thorium Nitrate, 400 ppm

Thorium nitrate solution was prepared by dissolving 0.952 g of $Th(NO_3)_4 \cdot 4 H_2O$ in distilled water and diluting to the mark in a 1 L volumetric flask.

Radium -226

A standard stock solution of Ra -226 was prepared by diluting a certified solution of 6.47 µCi of Ra -226 in 5 mls of 0.5 M HCI to 2.0 litres. The stock solution contained 16,175 pCi of Ra -226 per millilitre. Radium solutions of required concentrations were made by dilution of measured volumes of the stock solution.

Barium Nitrate

Barium nitrate weighed out and transferred to a 1 L volumetric flask. Distilled water was added to dissolve sample. The solution was then diluted to the mark. Three solutions were prepared, 0.01, 0.1 and 0.2 M barium nitrate. The respective weights of $Ba(NO_3)_2$ were 2.614 g, 26.14 g and 52.28 g.

Copper Nitrate

The copper nitrate solutions were prepared the same way as the barium nitrate solutions. The weights of $Cu(NO_3)_2 \cdot 2\frac{1}{2} H_2O$ used were 2.326 g, 23.26 g and 46.5 g per litre for 0.01 M, 0.1 M and 0.2 M solutions of copper respectively.

Ferric Nitrate Solutions

Solutions of ferric nitrate were prepared the same way as the barium nitrate solutions. The weights of $Fe(NO_3)_3 \cdot 9\ H_2)$ used were 4.04 g, 40.40 g and 80.80 g per litre for 0.01 M, 0.1 M and 0.2 M solution in ferric iron respectively.

Zinc Nitrate Solutions

Solutions of zinc nitrate were prepared the same way as the barium nitrate solutions. The weights of $Zu(NO_3)_2 \cdot 6\ H_2O$ used were 2.97 g, 29.75 g and 59.49 g per litre for 0.01 M, 0.1 M and 0.2 M solutions in zinc respectively.

Sodium Arsenate

Sodium arsenate solutions, 0.02 M, 0.2 M and 0.4 M, were prepared by dissolving 6.24, 62.4 and 124.8 g of $Na_2HAsO_4 \cdot 7\ H_2O$ in one litre of distilled water.

Sodium Sulphate

Sodium sulphate solution, 1 molar, was prepared by dissolving 142 g of Na_2SO_4 in 1 litre of distilled water.

2. Methods of Analysis

Determination of Radium

Instrumentation:

Canberra Multichannel Analyzer Model 8180
Preamp Model 2004
DC Power Supply
Detector Model PD-300-24-100M

Settings:

DC Voltage 35
Gain Coarse 10
Fine 8.62
Count Time 2000 sec.

Samples of barren liquors and column effluents were evaporated to near dryness, transferred to planchettes and further evaporated to dryness. The dried samples were analyzed for radium by counting alpha particles with the alpha spectrometer. The counts acquired with the samples were compared with the counts from standards and the quantity of radium calculated.

Determination of Thorium

Pipet 25 mls of aqueous sample into a 500 ml Ehrlenmeyer flask and dilute to 200 mls. Add 4 drops of Alizarin Red S indicator and adjust pH to 2.5 with ammonium hydroxide of nitric acid. Titrate solution with 0.025 M versene until pink in color begins to fade rapidly. Adjust the pH to 3.0 and continue titration. The end point is indicated when the pink color disappears giving a clear yellow color.

Calculation

$$\text{mg of Th/L} = \frac{232.1 \times V \times M}{\text{Sample Volume}} \times 1000$$

V = Volume of versene solution

M = Molarity of versene solution.

Determination of Metal Ions (Ba, Cu, Fe, Pb and Zn)

Instrument: Perkin Elmer Model 560, Atomic Absorption Spectrophotometer.

Conditions: Air - Acetylene

Ba - 553.6 mm

Cu - 324.7 mm

Fe - 248.3 mm

Pb - 283.3 mm

Zn - 213.9

To 10 ml aliquots of solution was added 1 ml of concentrated nitric acid in a 50 ml volumetric flask. The absorption obtained in the atomic absorption instrument was compared with the absorption of standard solutions and the metal iron concentration calculated.

The determination of barium required the addition of 2 mls of 25 g/100 ml KCI solution and 1 ml of concentrated nitric acid.

APPENDIX B

COPRECIPITATION DATA

TABLE B1

Coprecipitation of Lead and Zinc
As Arsenates

	A		B		C	
pH	Zn ppm	Pb ppm	Zn ppm	Pb ppm	Zn ppm	Pb ppm
1	2.8	97.0	27.2	97.7	54.9	108.2
2	2.9	97.0	26.7	101.1	54.6	102.9
3	2.9	90.1	27.1	6.2	56.9	2.0
4	2.6	61.2	24.8	0.14	54.2	0.16
5	2.6	53.4	24.6	0.14	51.6	0.16
6	2.7	50.0	23.3	0.28	23.8	<0.1
7	2.4	16.9	3.0	0.14	2.6	<0.1
8	2.4	20.9	1.2	1.5	1.6	0.3
9	1.8	2.6	1.3	3.0	6.2	5.3
10	0.02	0.14	2.8	8.7	5.9	5.8
11	0.12	0.86	7.6	27.0	42.3	63.3
12	1.6	4.3	3.8	12.8	9.6	18.4

Initial Concentrations:

	A	B	C
Zinc Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Lead Nitrate	4.8×10^{-4}	100 ppm Pb	100 ppm Pb

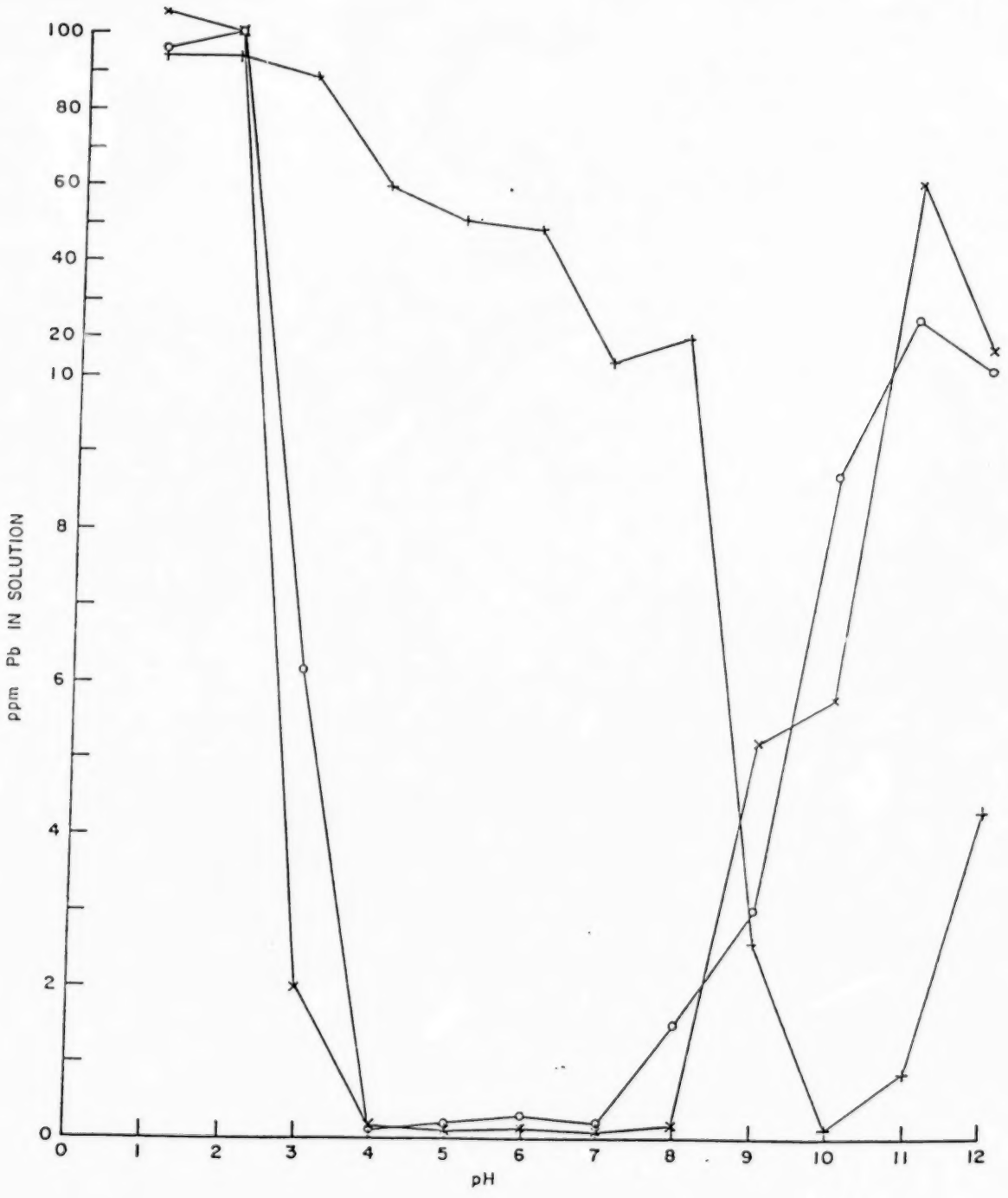

Fig. Coprecipitation of Lead with Zinc Arsenate as a function of pH

Initial concentrations :

Pb	100 ppm
Zn	0.01 M + ; 0.1 M o ; 0.2 M ×
Arsenate	0.02 M + ; 0.2 M o ; 0.4 M ×

TABLE B2

Coprecipitation of Lead and Copper
as Arsenates

	A		B		C	
pH	Pb ppm	Cu ppm	Pb ppm	Cu ppm	Pb ppm	Cu ppm
1	96.3	3.1	99.3	32.4	93.5	67.1
2	94.9	3.0	103.0	32.6	90.9	67.1
3	82.4	3.1	4.0	31.1	11.8	67.1
4	68.2	3.0	0.43	28.0	0.21	49.0
5	64.2	3.1	0.50	13.7	0.57	23.6
6	65.2	3.0	0.14	2.7	0.07	1.28
7	57.4	2.6	<0.01	<0.01	0.07	0.09
8	29.2	1.7	0.07	0.02	0.79	0.76
9	6.7	1.1	0.36	0.15	1.48	1.94
10	1.1	0.04	0.36	0.20	1.70	1.72
11	0.7	<0.01	0.50	<0 01	0.29	0.20
12	4.1	0.02	1.1	0.09	1.85	0.09

Initial Concentrations:

	A	B	C
Copper Nitrate	1×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Lead Nitrate	100 ppm Pb	100 ppm Pb	100 ppm Pb

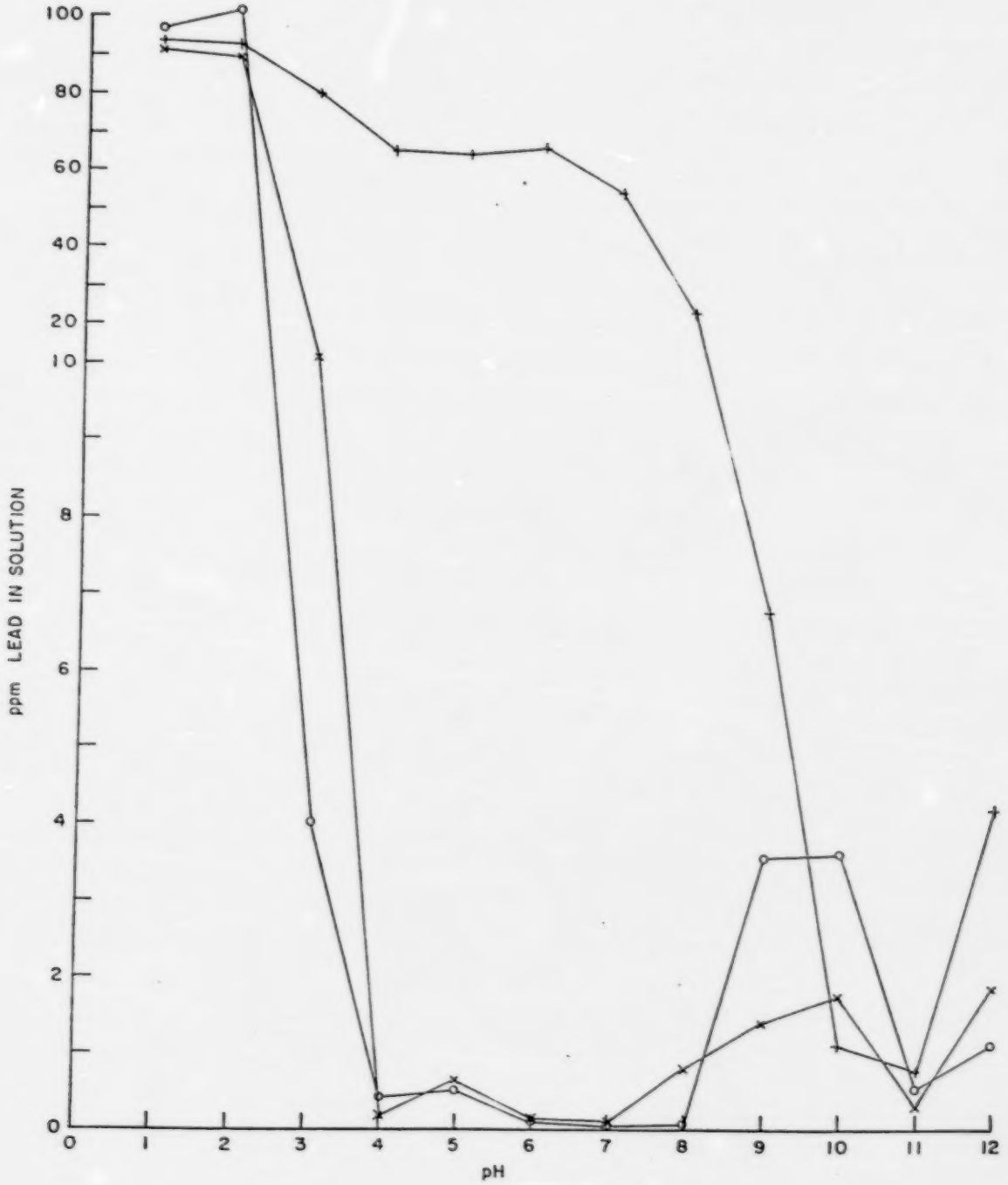

Fig. Coprecipitation of Lead with Copper Arsenate

Initial concentrations :

Pb 100 ppm

C. 0.01 M + ; 0.1 M o ; 0.2 M ×

Arsenate 0.02 M + ; 0.2 M o ; 0.4 M ×

TABLE B3

Coprecipitation of Lead and Barium
as Arsenates

	A		B		C	
pH	Ba ppm	Pb ppm	Ba ppm	Pb ppm	Ba ppm	Pb ppm
1	4.2	88.5	58.3	88.8	122.9	86.3
2	6.3	90.0	56.3	93.7	129.2	91.3
3	6.3	93.7	60.4	5 0	120.8	2.5
4	4.2	60.7	54.2	0.44	118.8	0.19
5	6.3	58.3	62.5	0.11	122.9	0.11
6	4.2	53.4	58.3	0.63	122.9	0.10
7	4.2	48.8	54.2	0.05	112.5	0.06
8	8.3	26.1	41.7	0.26	114.6	0.15
9	4.2	14.4	41.7	0.24	106.2	2.25
10	6.3	0.25	35.4	0.11	91.7	0.10
11	2.1	0.40	18.8	0.29	33.3	0.05
12	2.1	8.9	45.8	8.0	8.3	6.2

Initial Concentrations:

	A	B	C
Barium Nitrat	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Lead Nitrate	100 ppm Pb	100 ppm Pb	100 ppm Pb

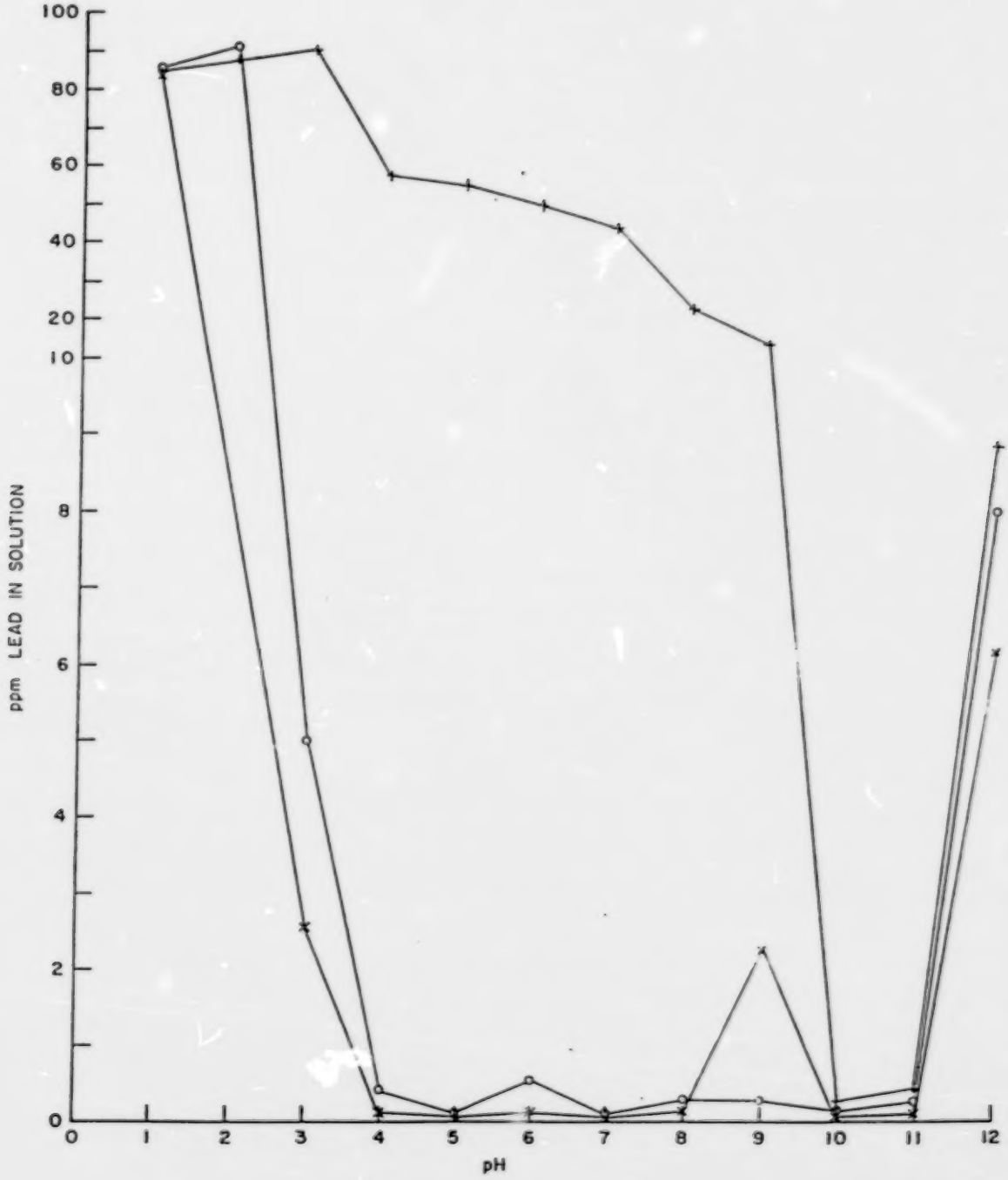

Fig. Coprecipitation of Lead with Barium Arsenate

Initial concentrations :

Pb 100 ppm

Ba 0.01 M + ; 0.1 M o ; 0.2 M ×

Arsenate 0.02 M + ; 0.2 M o ; 0.4 M ×

TABLE B4

Coprecipitation of Lead and Ferric Iron
As Arsenates

	A		B		C	
pH	Pb ppm	Fe ppm	Pb ppm	Fe ppm	Pb ppm	Fe ppm
1	6.9	0.3	91.0	25.0	97.5	50.6
2	7.4	0.3	91.6	23.5	97.5	45.9
3	7.1	0.3	36.5	<0.01	14.4	0.06
4	5.3	<0.01	2.7	0.06	0.56	<0.01
5	5.0	0.06	0.67	0.06	<0.01	0.3
6	5.6	0.06	<0.01	0.06	<0.01	<0.01
7	3.8	0.06	1.0	0.3	3.9	0.24
8	2.6	0.06	1.3	<0.01	7.2	0.88
9	1.2	0.06	3.0	0.65	6.7	1.5
10	0.3	<0.01	4.7	1.0	1.7	1.5
11	2.4	<0.01	41.0	10.7	45.2	43.5
12	<0.01	0.3	7.1	0.3	1.1	0-41

Initial Concentrations:

	A	B	C
Ferric Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Lead Nitrate	100 ppm Pb	100 ppm Pb	100 ppm Pb

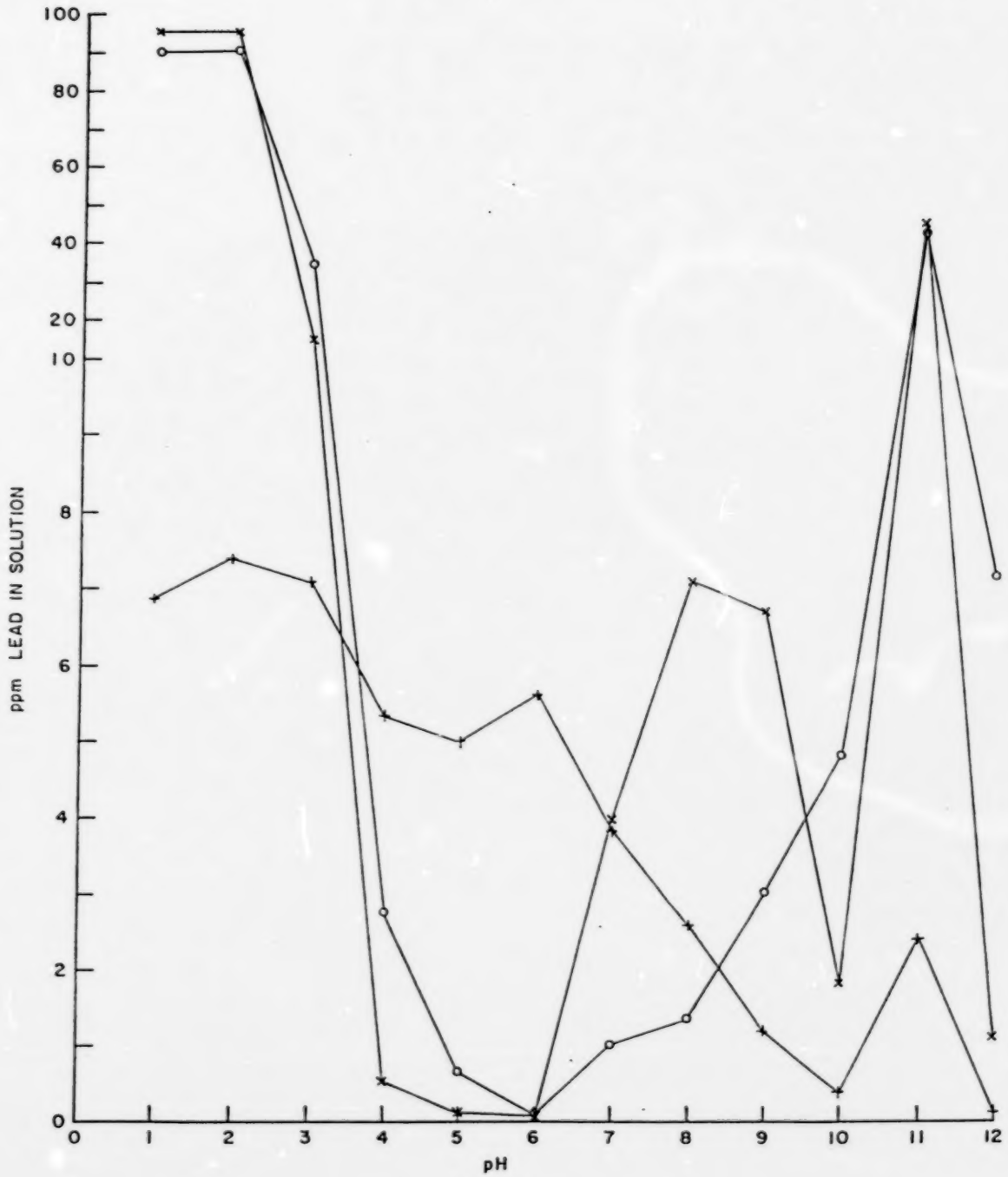

Fig. Coprecipitation of Lead with Ferric Arsenate

Initial concentrations :

Pb 100ppm

Fe 0.01 M + ; 0.1 M o ; 0.2 M x

Arsenate 0.02 M + ; 0.2 M o ; 0.4 M x

TABLE B5

Coprecipitation of Thorium and Zinc
as Arsenates

	A		B		C	
pH	Zn ppm	Th ppm	Zn ppm	Th ppm	Zn ppm	Th ppm
1	2.6	48.7	27.3	23.2	54.0	23.2
2	2.6	74.2	28.4	<0.01	54.7	<0.01
3	2.7	65.0	28.9	<0.01	54.7	<0.01
4	2.7	51.0	27.3	<0.01	48.0	<0.01
5	2.5	4.6	14.6	<0.01	35.0	<0.01
6	1.3	<0.01	1.1	<0.01	15.5	<0.01
7	0.49	<0.01	1.1	<0.01	1.4	<0.01
8	0.10	<0.01	7.2	<0.01	8.8	<0.01
9	0.13	<0.01	10.3	<0.01	18.4	<0.01
10	0.94	<0.01	7.6	<0.01	20.5	<0.01
11	0.42	<0.01	3.6	<0.01	17.3	<0.01
12	0.27	<0.01	5.0	<0.01	2.8	<0.01

Initial Concentrations:

	A	B	C
Zinc Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Thorium Nitrate	100 ppm Th	100 ppm Th	100 ppm Th

TABLE B6

Coprecipitation of Thorium and Copper as Arsenates

	A		B		C	
pH	Cu ppm	Th ppm	Cu ppm	Th ppm	Cu ppm	Th ppm
1	2.7	60.3	29.6	32.5	59.8	
2	3.0	71.9	33.0	<0.01	61.7	9.2
3	3.1	67.3	30.6	<0.01	60.8	<0.01
4	3.3	55.7	25.5	<0.01	49.1	<0.01
5	2.4	<0.01	6.8	<0.01	12.4	<0.01
6	0.09	<0.01	0.13	<0.01	0.65	<0.01
7	0.02	<0.01	<0.01	<0.01	0.22	<0.01
8	0.02	<0.01	1.3	<0.01	3.2	<0.01
9	0.02	<0.01	0.06	<0.01	2.9	<0.01
10	0.04	<0.01	0.17	<0.01	5.0	<0.01
11	0.02	<0.01	0.02	<0.01	0.87	<0.01
12	0.04	<0.01	0.06	<0.01	0.02	<0.01

Initial Concentrations:

	A	B	C
Copper Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Thorium Nitrate	100 ppm Th	100 ppm Th	100 ppm Th

TABLE B7

Coprecipitation of Thorium and Barium
as Arsenates

	A		B		C	
pH	Ba ppm	Th ppm	Ba ppm	Th ppm	Ba ppm	Th ppm
1	4.7	69.6	80.0	27.8	176.9	4.6
2	3.1	92.8	75.3	2.8	180.0	<0.01
3	10.9	74.2	76.9	<0.01	167.6	<0.01
4	7.8	53.3	80.0	<0.01	176.9	<0.01
5	9.4	7.0	73.3	<0.01	166.2	<0.01
6	7.8	1.0	65.6	<0.01	152.3	<0.01
7	6.3	<0.01	67.1	<0.01	141.5	<0.01
8	6.3	<0.01	61.5	<0.01	136.9	<0.01
9	4.7	<0.01	60.0	<0.01	156.9	<0.01
10	4.7	<0.01	53.8	<0.01	141.5	<0.01
11	<0.1	<0.01	40.6	<0.01	96.9	<0.01
12	<0.1	<0.01	3.1	<0.01	3.1	<0.01

Initial Concentrations:

	A	B	C
Barium Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Thorium Nitrate	100 ppm Pb	100 ppm Pb	100 ppm Pb

TABLE B8

Coprecipitation of Thorium and Ferric Iron as Arsenates

	A		B		C	
pH	Fe ppm	Th ppm	Fe ppm	Th ppm	Fe ppm	Th ppm
1	2.3	69.6	23.8	116.0	48.1	4.6
2	2.5	53.3	23.1	34.8	44.4	<0.01
3	2.3	97.4	19.4	<0.01	0.18	<0.01
4	2.4	48.7	0.06	<0.01	0.12	<0.01
5	0.06	<0.01	<0.01	<0.01	0.03	<0.01
6	0.06	<0.01	11.1	<0.01	2.6	<0.01
7	<0.01	<0.01	11.8	<0.01	8.8	<0.01
8	<0.01	<0.01	10.6	<0.01	4.8	<0.01
9	0.06	<0.01	12.9	<0.01	8.8	<0.01
10	0.06	<0.01	16.9	<0.01	17.5	<0.01
11	0.18	<0.01	8.8	<0.01	25.0	<0.01
12	0.18	<0.01	0.24	<0.01	0.35	<0.01

Initial Concentrations:

	A	B	C
Ferric Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Thorium Nitrate	100 ppm Th	100 ppm Th	100 ppm Th

TABLE B9

Coprecipitation of Radium and Zinc
as Arsenates

	A		B		C	
pH	Ra pCi	Zn ppm	Ra pCi	Zn ppm	Ra pCi	Zn ppm
1	25		31	36	13	70
2	81		37	38	14	67
3	56		16	39	8	78
4	60		21	39	6	66
5	81		15	34	16	65
6	50		23	37	2	17
7	79		8	4.1	0	4.4
8	35		21	36	9	24
9	18		38	36	8	41
10	3		14	33	5	61
11	5		2	2.6	4	5.1
12	6		3	7.4	0	1.5

Initial Concentrations:

	A	B	C
Zinc Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Radium 226	2500 pCi/L	2500 pCi/L	2500 pCi/L

TABLE B10

Coprecipitation of Radium and Copper
as Arsenates

	A		B		C	
pH	Ra pCi	Cu ppm	Ra pCi	Cu ppm	Ra pCi	Cu ppm
1	151		50	40.6	11	90.3
2	199		24	43.8	9	87.6
3	66		23	44.0	9	87.6
4	81		23	43.8	7	80.1
5	78		20	39.4	5	25.9
6	41		18	2.4	0	0.9
7	9		0	<0.01	0	0.5
8	8		0	<0.01	4	6.8
9	19		3	3.2	5	7.3
10	18		0	0.2	6	7.7
11	21		4	8.8	5	5.0
12	26		2	<0.01	0	<0.01

Initial Concentrations:

	A	B	C
Copper Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Radium 226	2500 pCi/L	2500 pCi/L	2500 pCi/L

TABLE B11

Coprecipitation of Radium and Barium
as Arsenates

	A		B		C	
pH	Ba ppm	Ra pCi	Ba ppm	Ra pCi	Ba ppm	Ra pCi
1		63	90.0	44	131.5	35
2		89	86.7	53	134.2	40
3		91	100.0	39	144.7	15
4		131	93.3	39	142.1	15
5		52	93.3	10	136.8	8
6		56	86.7	7	144.7	3
7		44	90.0	58	144.7	15
8		186	96.7	36	139.5	14
9		52	86.7	24	142.1	12
10		44	80.0	7	147.4	27
11		74	73.3	16	63.1	8
12		29	3.6	0	<0.01	0

Initial Concentrations:

	A	B	C
Barium Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Radium 226	2500 pCi/L	2500 pCi/L	2500 pCi/L

TABLE B12

Coprecipitation of Radium and Iron
as Arsenates

	A		B		C	
pH	Fe ppm	Ra pCi	Fe ppm	Ra pCi	Fe ppm	Ra pCi
1		83	48.6	44	77.3	42
2		166	16.6	65	28.8	21
3		5	0.2	35	<0.01	14
4		37	<0.01	35	<0.01	9
5		115	1.2	7	<0.01	9
6		9	0.6	15	<0.01	5
7		3	18.1	11	27.5	21
8		111	27.7	0	40.9	0
9		80	35.1	55	68.0	14
10		6	44.6	31	71.5	14
11		23	36.1	12	65.5	0
12		44	2.7	18	2.5	0

Initial Concentrations:

	A	B	C
Ferric Nitrate	5×10^{-5} M	5×10^{-4} M	1×10^{-3} M
Sodium Hydrogen Arsenate	1×10^{-4} M	1×10^{-3} M	2×10^{-3} M
Radium 226	2500 pCi/L	2500 pCi/L	2500 pCi/L

TABLE B13

Coprecipitation of Lead
From a 0.75 M Sodium Sulphate Solution

	Zn		Cu		Ba		Fe	
pH	Zn ppm	Pb ppm	Cu ppm	Pb ppm	Ba ppm	Pb ppm	Fe ppm	Pb ppm
1	72.1	7.6	85.5	7.5	<0.01	0.56	24.0	5.0
2	68.7	8.2	80.0	8.1	1.3	1.2	25.4	7.8
3	65.8	10.0	83.6	8.3	1.3	0.67	4.7	8.0
4	69.8	9.7	76.8	10.2	2.5	1.3	0.6	7.1
5	68.1	0.57	72.7	2.7	1.3	0.8	0.5	2.8
6	62.3	1.8	8.8	0.33	1.3	1.0	0.7	0.8
7	47.3	0.43	0.9	0.33	1.3	0.56	0.9	0.8
8	4.8	0.29	0.12	0.11	2.5	0.9	1.8	1.4
9	0.36	0.14	0.1	0.22	1.3	0.4	3.1	2.9
10	13.6	3.1	0.07	0.33	2.5	2.0	4.0	7.1
11	0.9	1.8	0.09	0.11	3.8	3.4	0.6	0.3
12	18.8	0.7	0.1	1.9	<0.01	0.7	0.5	0.4

Initial Concentrations:

Nitrates of Coprecipitants	1×10^{-3} M
Sodium Hydrogen Arsenate	2×10^{-3} M
Lead Nitrate	100 ppm pb

TABLE B14

Coprecipitation of Thorium
From a 0.75 M Sodium Sulphate Solution

	Zn		Cu		Ba		Fe	
pH	Zn ppm	Th ppm	Cu ppm	Th ppm	Ba ppm	Th ppm	Fe ppm	Th ppm
1	62.3	<0.01	66.1	<0.01	<0.01	<0.01	40.0	<0.01
2	61.2	<0.01	68.3	<0.01	<0.01	<0.01	43.6	<0.01
3	61.2	<0.01	66.1	<0.01	<0.01	<0.01	4.4	<0.01
4	62.9	<0.01	66.7	<0.01	<0.01	<0.01	0.6	<0.01
5	61.2	<0.01	60.6	<0.01	<0.01	<0.01	0.4	<0.01
6	49.0	<0.01	8.9	<0.01	<0.01	<0.01	0.5	<0.01
7	43.2	<0.01	0.4	<0.01	<0.01	<0.01	0.7	<0.01
8	5.9	<0.01	0.2	<0.01	3.5	<0.01	0.8	<0.01
9	1.2	<0.01	0.1	<0.01	3.5	<0.01	1.3	<0.01
10	0.1	<0.01	0.1	<0.01	1.8	<0.01	1.1	<0.01
11	0.3	<0.01	0.1	<0.01	3.5	<0.01	0.6	<0.01
12	3.8	<0.01	0.1	<0.01	1.8	<0.01	0.6	<0.01

Initial Concentrations:

Nitrates of Coprecipitants	1×10^{-3} M
Sodium Hydrogen Arsenate	2×10^{-3} M
Thorium Nitrate	100 ppm Th

TABLE B15

Coprecipitation of Radium
From a 0.75 M Sodium Sulphate Solution

	Ba		Fe	
pH	Ba ppm	Ra pCi	Fe ppm	Ra pCi
1	<0.1	18	69.1	13
2	<0.1	7	63.8	6
3	<0.1	6	2.0	4
4	<0.1	6	2.0	2
5	<0.1	3	2.0	0
6	<0.1	1	4.0	4
7	<0.1	0	8.0	2
8	<0.1	0	10.0	4
9	<0.1	0	0.5	0
10	<0.1	1	4.0	2
11	<0.1	0	0.5	0
12	<0.1	0	0.5	0

Initial Concentrations:

Ferric Nitrate	1×10^{-3} M
Barium Nitrate	1×10^{-3} M
Sodium Hydrogen Arsenate	2×10^{-3} M
Radium	2500 pCi/L